PRACTICAL GAMMA SPECTROSCOPY ASSAY TECHNIQUES FOR LARGE VOLUME LOW-LEVEL WASTE BOXES

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ABSTRACT

A study was conducted at the Los Alamos National Laboratory (LANL) to evaluate the performance of the SNAP™ (Spectral Nondestructive Assay Platform) analytical software for measurements of known standards in large metal waste boxes (2.5 m^3 volume). The trials were designed to test the accuracy and variance of the analytical results for low-density combustible matrices and higher-density metal matrices at two discrete gamma-ray energies: 121.78 keV and 411.12 keV. For both matrix types the measurement method that produced the most accurate results with the lowest associated standard deviation involved combining four individual measurements taken at the geometric center of each of the box's four vertical sides. With this method the overall bias and the standard deviation amongst 24 individual results for the 121.78 keV and 411.12 keV gamma rays were as follows: 3.38% (± 20.19%) and 3.68% (± 15.47%) for the combustible matrix and 37.88% (± 67.64%) and 9.38% (± 33.15%) for the metal matrix. The persistent positive bias from measurements of the metal box is believed to be a result of a non-homogenously distributed matrix.

INTRODUCTION

The Solid Waste Operations (SWO) group at LANL performs radioactive waste characterization measurements on a variety of waste items using portable high-purity germanium (HPGe) detectors. One common waste container used at LANL is a large metal box known as the "B25 Box". The B25 is a 2.5 m^3 (90 ft^3) rectangular box measuring 47 inches high by 72 inches wide by 45.5 inches deep. B25s are used for the disposal of low-level waste (LLW) and are loaded with a large variety of waste materials and contaminated with many different radionuclides. When the contaminants include transuranic (TRU) radionuclides the assay method must be able to confidently detect and quantify those isotopes below the TRU concentration cutoff of 100 nCi/g. Once the raw data has been collected, SWO uses the SNAP™ analytical software from Eberline Services to produce assay results for the radionuclides that were detected.

The SNAP™ gamma spectroscopy analytical software incorporates the raw data acquired from portable HPGe detectors and applies it to a physical model of the item to be assayed. All the elements normally associated with a system's calibration factor are mathematically calculated to produce a method-specific 'calibration' each time: the item-to-detector geometry, the matrix attenuation losses of gamma-rays headed towards the detector, attenuation losses from other shields in the gamma-ray path (e.g., container wall), the HPGe intrinsic efficiency, and the gamma-ray emission probability per decay. This mathematical modeling approach using portable detectors allows the analyst unlimited flexibility to make *a priori* decisions regarding the measurement protocol (e.g., item-to-detector distance and position, count time, etc.) to ensure measurement results will meet pre-defined expectations (e.g., < 100 nCi/g sensitivity).

Like all analysis techniques, there are limitations to the accuracy that can be achieved with this approach. Furthermore, the accuracy of the final result is closely linked to the measurement technique that was used in the assay count. For example, one would not expect the assay result from a single 10 minute measurement on just one side of the B25 to be as consistently accurate as an assay result derived from four 10-minute counts with one count taken from each of the four vertical sides. There are a variety of measurement techniques that can be employed with a portable HPGe detector that is easily moved around the different sides of a B25 box. The two questions that we attempt to address in this study are: 1) what are the optimal measurement and modeling techniques to employ with portable HPGe detectors for the assay of B25 boxes, and 2) what level of accuracy and consistency can we expect for radionuclides with relatively low-energy gamma-ray emissions (122 keV) and medium-energy gamma-ray emissions (411 keV) in a B25 box?

METHODS

Two B25 boxes were configured with mock matrices of common materials seen in LANL wastes: combustibles and metals. The combustible matrix consisted of 227.7 kg (502 lbs) of bags with shredded paper and miscellaneous plastics. The metal matrix consisted of 735.7 kg (1622 lbs) of various sized pieces of carbon steel. One quadrant of the B25 box contained two vertical rows of four evenly spaced PVC tubes. Two Eu-152 sources (0.5 mCi and 1.0 mCi) were placed at selected heights within each of the tubes (heights above the bottom were 4", 12", and 20") . We assumed that if the matrix distribution was reasonably uniform that the source placement in a single quadrant would mirror the placement in the other quadrants. Within the quadrant we selected 24 unique source locations that were designed to closely mimic a 'uniform' source distribution. See figure 1 for a description of the source locations.

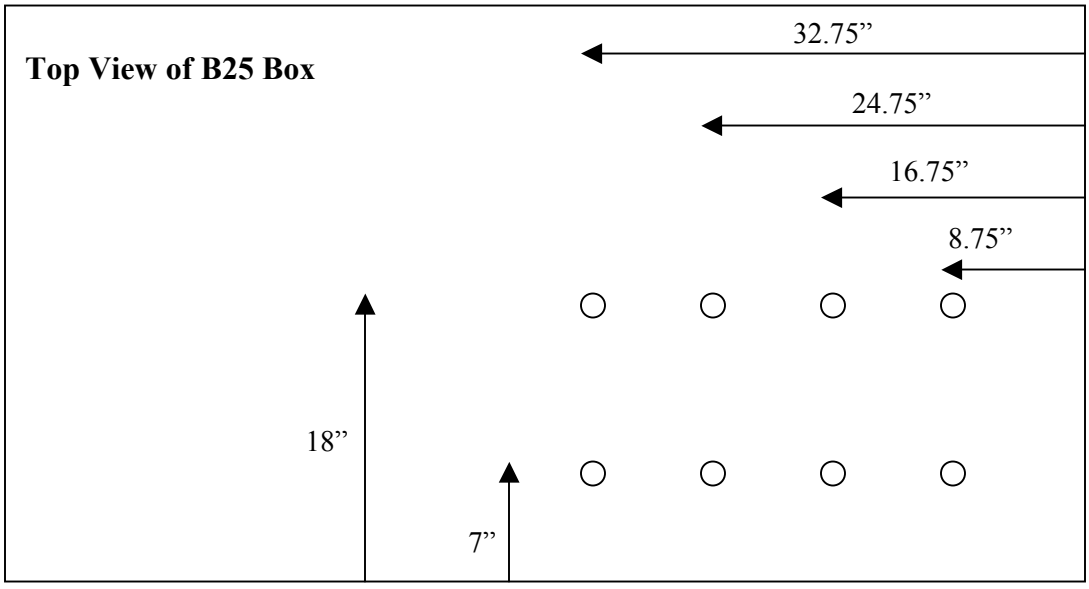


Figure 1. Source locations within the B25

All measurements were performed with portable HPGe detectors that were mounted on a height adjustable cart (the carbon steel matrix B25 was measured with a 28% relative efficiency detector, while the paper matrix was measured with a 45% detector). A portable digital signal processor and laptop computer were placed on another cart adjacent to the detector. While the detector crystal was shielded on the sides with 5.08 cm (2 in) of lead, it was not collimated inside the lead collar. Instead, the front face of the detector crystal was flush with the edge of the lead collar. In this manner the full front surface area of the crystal is visible to the entire volume of the B25 box being measured during the entire count. This lack of collimation increases the total fraction of gamma-rays emitted from the B25 box that can be seen by the detector, thus lowering the inherent limits of detection for any given radionuclide. Figure 2 illustrates the detector and shielding configuration.

Fig. 2. Detector and shielding configuration

The detector was vertically centered at eight distinct locations around the B25 box at a distance of 61 cm (24 inches) from the side (see figure 3 for the detector locations). For each matrix material, a single count was performed at each of the eight detector locations for each of the 24 unique source locations in the box for a total of 192 counts. The count times were determined by the need to obtain reasonable count statistics in the two gamma-ray peaks of interest (122 keV and 411 keV). Normally a brief 300 sec count was adequate to ensure a minimum of 1000 net counts were present in each peak. However, count times were extended whenever necessary (which was frequently the case for the carbon steel matrix) and data were sometimes deemed acceptable when just 200 – 300 net counts were present.

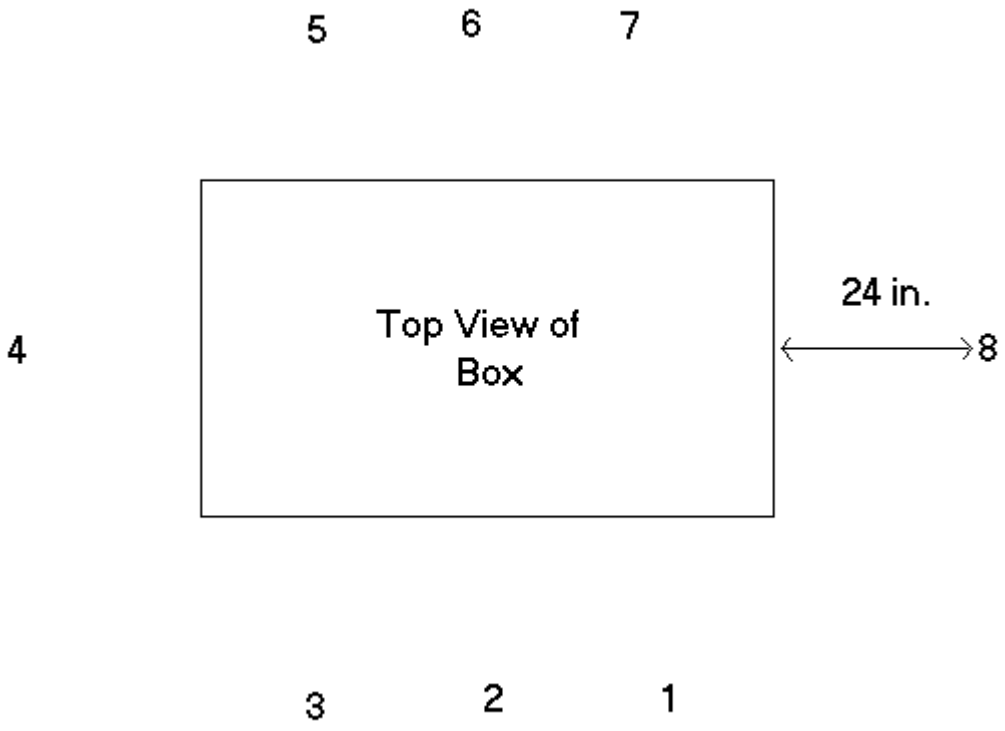


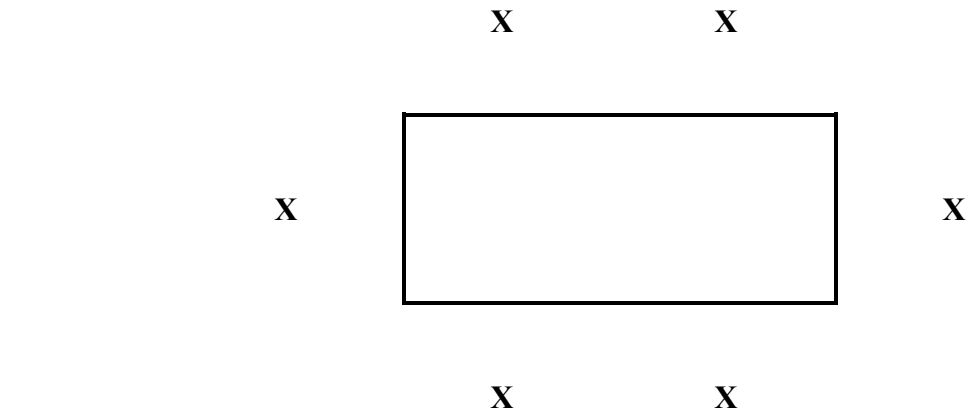
Figure 3. Eight detector positions during measurement trials

Several detector placement combinations were determined as possible modeling techniques for the B25s. We decided to analyze assay results using the following combinations of different detector positions: 1) the detector placed on two opposing sides, 2) the detector centered on all four vertical sides (excluding the top and bottom), and 3) a special six-count configuration on four sides. A visual depiction of the possible measurement combinations is presented in figure 4.

Two different types of modeling were used for the combinations which involved counts on all four sides. One method was to model the B25 as having it's actual dimensions (47 x 72 x 45.5) with the long side described as facing the detector. However, in reality the short sides are also facing the detector for part of the count. Therefore, a second modeling technique was evaluated in which the B25 is described as a square (47 x 57.24 x 57.24) having the same volume as the actual B25.

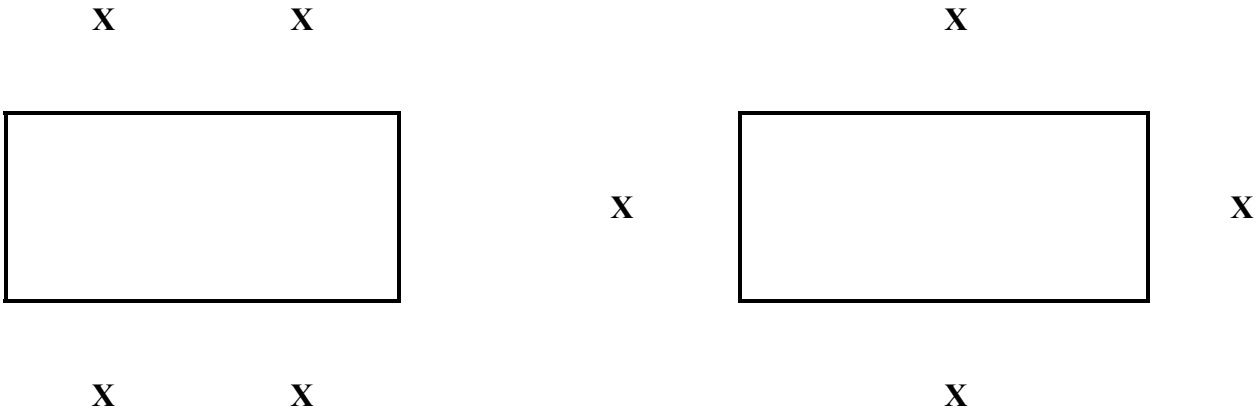
The calculated assay values for the Eu-152 source in the boxes were compared to a pre-trial calibration assay of the source. Eu-152 was chosen for the trials because it emits several gamma rays over a broad range of energies, including two gamma rays which are very close in energy to two primary gamma rays from Pu-239: a 121.78 keV gamma (Pu-239 has a 129.29) and a 411.12 keV gamma (Pu-239 has a 413.69). Because the two Eu-152 gammas are so close in energy to the Pu-239 gammas it can be expected that Pu-239 assays will have similar results in terms of accuracy and variance.

6 COUNT ASSAY OPTION



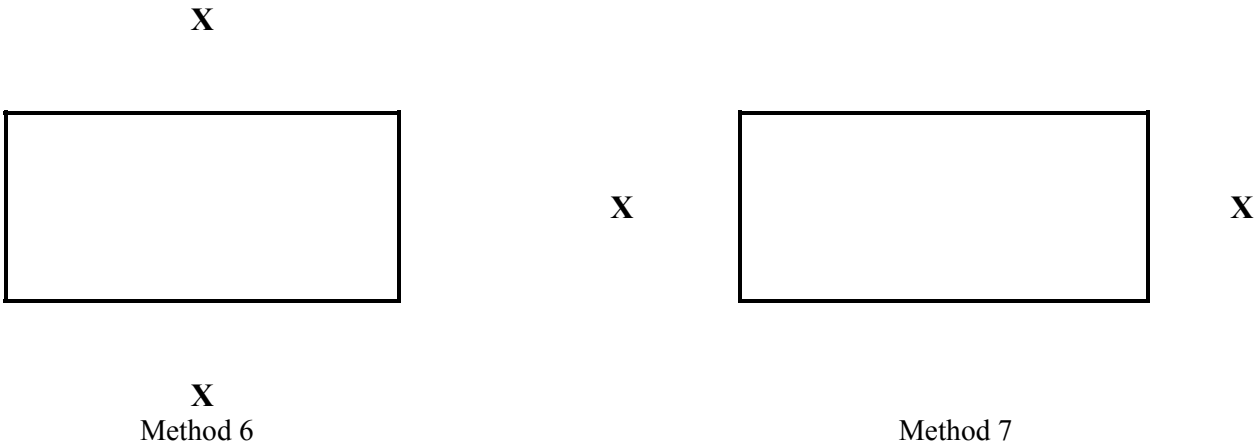
Methods 1 & 2

4 COUNT ASSAY OPTIONS



Method 3 Methods 4 & 5

2 COUNT ASSAY OPTIONS



Method 6 Method 7

Figure 4. Assay counting options.

RESULTS

The SNAP results vs. the known value for the combustible matrix B25 are presented in Table 1 and the SNAP results vs. the known value for the metal matrix B25 are presented in Table 2. The percent bias results evaluate the overall bias between the known source activity and the calculated activity averaged over all 24 source positions. The percent relative standard deviation (%RSD) results are a key indicator of the expected range of measurement results amongst the 24 unique measurement positions. Ideally, the best results will have a small overall measurement bias (e.g., < 10%) and a small %RSD (e.g., < 50%).

A few expected trends are readily observable in the results. The %RSD is largest when counts are made from just two sides of the box and smallest when counts are made from all four sides (regardless of matrix). The %RSD is also smallest for results derived with the higher-energy gamma at 411 keV vs. the low-energy gamma at 122 keV and it is also smaller for the lower density combustible matrix compared to the higher density metal matrix. The most accurate results (i.e., smallest % bias) in the metal matrix box are seen with the 411 keV gamma ray, while both gammas (122 keV and 411 keV) are seen to produce comparably accurate results in the combustible matrix box.

The best overall method in terms of measurement positions and modeling techniques for both matrices was method number 4. In this method a single count is taken from the geometric center of each of the four vertical sides of the box and then the box is modeled as a square having the same volume as the actual B25 (dimensions are input as 47" height x 57.24" width x 57.24" depth). Methods number 1 and 5 also produced reasonably good results for both matrix types. Method number 1 also models the B25 box as a square, but it involves a total of six measurement positions (2 on each long side and one on each short side). Method number 5 models the B25 as a rectangle and includes just one count from the geometric center of each of the four sides. Another noteworthy finding, though unexpected, was the persistent positive bias in the measurement results for the metal matrix box. This bias was present regardless of the combination of detector positions used, the modeling method employed, or the gamma ray energy used in the assay.

Table I: SNAP Results Vs. the Known Value for Combustible Matrix B25 Box

Method No.	Method Description	Percent Bias at 122 keV	%RSD at 122 keV	Percent Bias at 411 keV	%RSD at 411 keV
1	6 counts (square box)	0.09%	22.41%	1.16%	17.13%
2	6 counts (rectangular box)	-11.60%	22.13%	-10.24%	16.92%
3	4 counts (2 ea long side)	23.93%	35.43%	21.30%	28.12%
4	4 counts (square box)	3.38%	20.19%	3.68%	15.47%
5	4 counts (rectangular box)	-9.46%	20.20%	-8.76%	15.49%
6	2 counts (1 ea long side)	4.87%	45.72%	5.88%	36.47%
7	2 counts (1 ea short side)	7.90%	58.24%	7.54%	46.79%

Table II: SNAP Results Vs. the Known Value for Metal Matrix B25 Box

Method No.	Method Description	Percent Bias at 122 keV	%RSD at 122 keV	Percent Bias at 411 keV	%RSD at 411 keV
1	6 counts (square box)	53.24%	85.09%	11.07%	43.24%
2	6 counts (rectangular box)	66.74%	85.14%	9.27%	43.25%
3	4 counts (2 ea long side)	273.48%	161.99%	52.27%	52.87%
4	4 counts (square box)	37.88%	67.64%	9.38%	33.15%
5	4 counts (rectangular box)	49.40%	67.62%	7.49%	33.13%
6	2 counts (1 ea long side)	146.69%	87.17%	52.23%	54.82%
7	2 counts (1 ea short side)	5.53%	130.75%	4.25%	82.76%

DISCUSSION

Many of the results in this study were consistent with our expectations. We expected to see a lower variance in the overall results when four sides of the box were measured as opposed to just two sides, and we did (methods 1 thru 5 compared to methods 6 and 7). The greatest variance was seen when we based the assay on two measurements of the short sides of the box. In this orientation the box has a depth of 72 inches and the variation in the actual distances gamma rays must travel to escape the matrix is the greatest.

We also expected to see a lower variance and greater accuracy for the results on the low-density combustible matrix compared to the results for the higher-density metal matrix. The results confirmed this expectation as well. Finally, we expected to see a lower variance and greater accuracy for results based on the 411 keV gamma ray compared to results based on the 122 keV gamma ray. In both matrices the variance in results was lowest for the 411 keV gamma, and in the metal matrix the overall bias was lowest for the 411 keV gamma as well. However, in the combustible matrix box there was no significant difference in the overall bias between results based on the 122 keV gamma compared to those based on the 411 keV gamma. We find this to be a reassuring result and believe it to be related to two key factors: 1) the combustible matrix was indeed uniformly distributed in the B25 box (thus matching the assumption of the SNAP modeling algorithm) and 2) the corrections for source geometry and matrix attenuation losses in the software are properly performed.

The significant positive bias in the results for the metal matrix box was not expected. Upon closer examination of individual measurement data we believe there is a logical explanation for this. Although the total mass of carbon steel was 735.7 kg (1622 lbs), the actual volume displaced by the matrix material was just 3.7% of the total volume in the box. In other words, the vast majority of the box volume (96.3%) is filled with air instead of steel. Because of this it became apparent that in some of the source positions, particularly those in the front row of tubes,

the Eu-152 gammas were encountering no matrix material whatsoever on their path towards the HPGe detector. Therefore we observed a significantly higher count rate in these cases than would be expected with a truly uniformly distributed matrix and a persistently high bias in our assay results. With respect for segregating TRU level wastes from LLW this result is not too disconcerting. It means that there is an increased likelihood that we would incorrectly declare LLW as TRU waste, which from the regulatory standpoint, is less of faux pas than declaring TRU waste as LLW.

REFERENCES

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